Surface effects in a semiconductor photonic nanowire and spectral stability of an embedded single quantum dot

Inah Yeo,[1] Nitin S. Malik,[2] Mathieu Munsch,[2] Emmanuel Dupuy,[2] Joël Bleuse,[2] Yann-Michel Niquet,[3] Jean-Michel Gérard,[2] Julien Claudon,[2, a)] Édouard Wagner,[1] Signe Seidelin,[1] Alexia Auffèves,[1] Jean-Philippe Poizat,[1] and Gilles Nogues[1, b)]

[1)] *CEA/CNRS/UJF joint team "Nanophysics and Semiconductors", Institut Néel-CNRS, BP 166, 25 rue des Martyrs, 38042 Grenoble Cedex 9, France*

[2)] *CEA/CNRS/UJF joint team "Nanophysics and Semiconductors", CEA/INAC/SP2M, 17 rue des Martyrs, 38054 Grenoble cedex 9, France*

[3)] *Laboratoire de Simulation Atomistique, CEA/INAC/SP2M, 17 rue des Martyrs, 38054 Grenoble cedex 9, France*

(Dated: 19 December 2011)

We evidence the influence of surface effects for InAs quantum dots embedded into GaAs photonic nanowires used as efficient single photon sources. We observe a continuous temporal drift of the emission energy that is an obstacle to resonant quantum optics experiments at the single photon level. We attribute the drift to the sticking of oxygen molecules onto the wire, which modifies the surface charge and hence the electric field seen by the quantum dot. The influence of temperature and excitation laser power on this phenomenon is studied. Most importantly, we demonstrate a proper treatment of the nanowire surface to suppress the drift.

PACS numbers: 78.55.-m,78.67.Uh,85.60.Jb,81.65.-b

[a)]Electronic mail: julien.claudon@cea.fr

[b)]Electronic mail: gilles.nogues@grenoble.cnrs.fr

Efficient nanophotonic devices like single photon sources require to funnel a large fraction β of the spontaneous emission (SE) of a single emitter into a single optical mode. This situation offers an ideal platform for quantum optics and quantum information processing experiments[1,2]. In this context, photonic semiconducting nanowires (PW) embedding a single quantum dot (QD) have emerged as appealing systems[3–5]. These monomode waveguides made of a high refractive index material, offer a tight lateral confinement of the guided mode while simultaneously screening all the other transverse modes. Hence they ensure an efficient SE control over a broad wavelength range ($\beta \geq$90% over a bandwidth exceeding 100 nm at λ =950 nm)[3,6]. Moreover the far-field outcoupling efficiency of the guided mode can reach unity by proper engineering of the wire ends[3]. Following this strategy, an on-demand single-photon source with a record-high brightness was demonstrated[4].

However, miniaturization of photonic devices enhances surface effects, inducing for example non radiative surface recombinations which affect the emission properties of semiconducting nanowires[7,8]. In the case of PWs, single-mode operation imposes a wire diameter $d \lesssim \lambda/n$, where n is the index of refraction of the material. For GaAs PWs the QD is thus located at distances not larger than 100 nm from sidewalls. It was shown in Ref. 6 that the almost perfect QD radiative yield is preserved in PWs with d as small as 200 nm. Photon correlation experiments showed no bunching in the 1–100 ns temporal range[4], a strong evidence of absence of blinking at this timescale. Nevertheless, the spectral stability of the PWs, crucial for resonant quantum optics experiments, has not been investigated so far. In this letter, we perform high resolution spectroscopy and show that the excitonic emission line of the QD undergoes a continuous energy drift. We discuss the possible origin of the phenomenon, which we attribute to oxygen adsorption on the wire sidewalls, and demonstrate a way to circumvent this problem with a proper surface treatment.

The device fabrication starts from a planar structure grown by molecular beam epitaxy on a GaAs wafer. A single layer of InAs self-assembled QDs (areal density 300 μm^{-2}) is located in a GaAs matrix (residual positive doping $p = 10^{16}$ cm^{-3}). The ensemble luminescence peaks at 920 nm (50 nm inhomogeneous broadening). PWs are defined with a top-down approach, using e-beam lithography and dry plasma etching ($Ar-SiCl_4$ plasma). A careful control of the etching leads to a conical wire geometry in order to control the optical mode transverse profile along the wire. A series of PWs with top diameters varying by a fine 10 nm step were fabricated. The actual wire diameter at the dot location d_{QD} is measured with a $\sim$10 nm

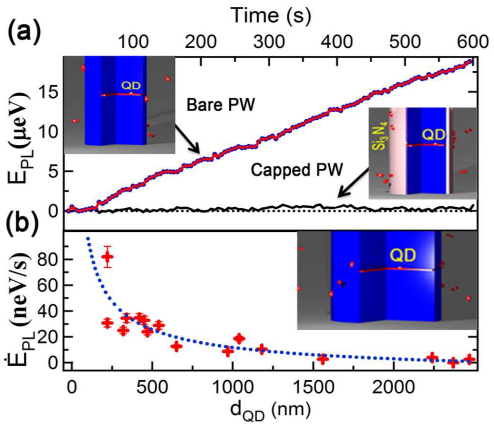

FIG. 1. (Color online) (a) $E_{\rm PL}$ versus time for a QD in a bare PW (blue line) and in a capped PW (black line). (b) $\dot{E}_{\rm PL}$ for QDs in bare PWs versus wire diameter

accuracy by electron microscopy. A wire with a diameter $d_{\rm QD}$ =200 nm typically contains 10 randomly located QDs. As surface passivation helps in reducing surface effects[8], we start from the previous device to fabricate surface capped-PWs. The oxide layer is removed by wet chemical etching. GaAs dangling bonds at the surface are saturated by $(NH_4)_2S$ in order to reduce the density of surface traps. Finally a 10 nm-thick layer of Si_3N_4 is deposited over the surface.

The sample is mounted on a cold finger cryostat with optical access. QDs are excited through a 0.75 NA microscope objective. Standard cw diode or Ti:Sa laser excite the QDs. Bandgap (BG) excitation at 1.52 eV creates electron-hole pairs in the bulk GaAs matrix. Wetting layer (WL) excitation (1.49 eV) only excites carriers in the continuum absorption band of the InAs monolayer close to the QDs. Finally quasi-resonant p-shell excitation (1.42 eV) excites discrete QD states. The light emitted by the recombination of the QD lowest energy states is recollected by the same lens and sent to a 1.5 m focal length spectrometer (12 μeV spectral resolution at 1.38 eV). A CCD camera is placed at the output of the spectrometer. Its pixel size (20 μm) corresponds to an energy step comparable to the resolution. Successive QD photoluminescence (PL) spectra are recorded and fitted with a Lorentzian function. Considering the signal to noise ratio for the data, we estimate the statistical uncertainty on the peak position $E_{\rm PL}$ to be 0.1 μeV.

Figure 1(a) presents the PL peak position $E_{\rm PL}$ versus time for a single QD in a $d_{\rm QD}$ =370 nm bare PW at 3.5 K. The quantum dot is excited by pumping the WL at

a power $P/P_{\rm sat} = 0.2$ ($P_{\rm sat}$ is the saturation power of the QD). A regular drift $\dot{E}_{\rm PL}$ of the emission energy towards higher frequencies is observed (blue drift). A linear fit of the data of Fig. 1(a) gives $\dot{E}_{\rm PL} \simeq$30 neV/s. At 3.5 K, $\dot{E}_{\rm PL}$ remains constant over 8 hours. Over this period of time, PL intensity does not change significantly, its linewidth remains limited by the resolution of the spectrometer and lifetime measurements show a small decrease from 1.65 to 1.58 ns. Warming up to room temperature and pumping on the cryostat resets the emission energy to its initial lower value. A systematic study of many PWs with different diameters [Fig. 1(b)] reveals that all QDs are affected by a blue drift whose amplitude decreases with increasing diameter. Finally the same experiment with QDs in capped PWs shows no drift (Fig. 1(a), $\dot{E}_{\rm PL} \leq$0.7 neV/s).

Previous observations clearly point towards surface effects. Dry etching of PWs during their fabrication creates a large density of surface traps[7] $n_s \gtrsim 10^{12}$ cm^{-2}. For GaAs, the energy of those traps is in the middle of the gap ($\Phi \equiv E_{\rm trap} - E_{\rm VB} =$0.7 eV)[9]. If the density of states at this energy is large enough, the Fermi level is pinned and hence the energy bands are bended close to the surface. Band bending is accompanied by a surface built-in electric field and a positive charging of the surface for p-doped semiconductors. Assuming a planar interface, the depletion length over which the bands are shifted is $W = \sqrt{\epsilon\epsilon_0\Phi/qp}$, where $\epsilon = 12.9$ is the dielectric constant of GaAs, ϵ_0 the vacuum permittivity and q the electron charge. For $p = 10^{16}$ cm^{-3}, $W =$300 nm ($> d_{\rm QD}$). Hence the region of the PW containing the dots is fully depleted and an electric field is present over its entire cross-section, explaining why all studied QDs experience a drift. We have confirmed this fact with numerical simulations. It shows that the field inside the wire is proportional to the surface charge. Due to the conical shape of the PW a small and almost constant $F_\parallel =$1.4 kV/cm component of the electric field exists along the axis of the wire. The radial electric field $F_\perp$ ranges from 0 kV/cm on the PW axis to $-$26 kV/cm at the surface. An embedded InAs QD subjected to this field experiences a Stark energy shift[10,11] $\Delta E_{\rm Stark} \approx -d_\parallel F_\parallel - \alpha_\perp F_\perp^2$, with $d_\parallel \approx$40 μeV$\cdot$kV^{-1}cm and $\alpha_\perp \approx$4 μeV$\cdot$kV^{-2}cm^2. We note that a blue drift implies a reduction of the electric field, hence a diminution of the positive net charge at the surface of the PW.

We attribute this effect to the physisorption of O_2 onto the surface leading to the creation of acceptor-like surface states that capture one electron[12–14]. The reset of $E_{\rm PL}$ to a low value after warming up to room temperature implies a reversible physical phenomenon and not an

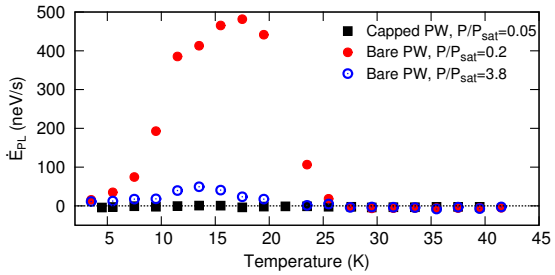

FIG. 2. (Color online) $\dot{E}_{\rm PL}$ as a function of T for excitation power $P/P_{\rm sat} = 0.2$ (•) and $P/P_{\rm sat} = 3.8$ ($\odot$) in a bare PW and for $P/P_{\rm sat} = 0.05$ in a capped PW (■)

irreversible chemical surface reaction. The full circles on Fig. 2 represent the dependence of $\dot{E}_{\rm PL}$ as a function of T between 4 and 45 K in the same conditions as in Fig. 1(a). A ten-fold increase occurs from 4 to 20 K before the drift dramatically drops to values very close to 0 above 30 K. This observation is in good agreement with previous studies on the adsorption of O_2 on GaAs, which evidenced an adsorption peak around 20 K[14,15]. The same experiment for a capped PW (full squares in Fig. 2) shows that $\dot{E}_{\rm PL}$ remains below 4 neV/s over the whole temperature range, proving the drift cancellation by the capping. The temperature dependence of $\dot{E}_{\rm PL}$ under high excitation power ($P/P_{\rm sat}$ =3.8; open circles in Fig. 2) exhibits the same characteristic peak. Its maximum value is shifted to lower temperature by ∼4 K and is dramatically smaller than for low power excitation. The pump laser can affect the PW in different ways: it can locally warm up the PW (explaining in particular the peak shift at high power in Fig. 2), photocreated carriers can screen the electric field[9] or change the adsorption rate of O_2.

To better understand the phenomenon, we performed systematic power studies at T =4 K. Figure 3(a) presents a typical experimental run: the drift $\dot{E}_{\rm PL}$ is determined from measurement of the emission energy over 5 min at different power values. Here, we also observe sudden shifts of the emission energy $\Delta E_{\rm PL} < 0$ when the power is increased. We attribute it to a local warming of the PW ($T_{\rm PW} \geq T$) leading to the usual quadratic shift of the bandgap. For p-shell excitation, the amplitude of $\Delta E_{\rm PL}$ is below the fitting uncertainty on the peak position (0.1 μeV). In the other two cases, $|\Delta E_{\rm PL}|$ increases linearly with power. Its magnitude is larger for BG excitation compared to WL excitation. Similar values have been observed for many QDs in PWs of similar size, capped or not.

The influence of power on $\dot{E}_{\rm PL}$ crucially depends on the excitation conditions (see

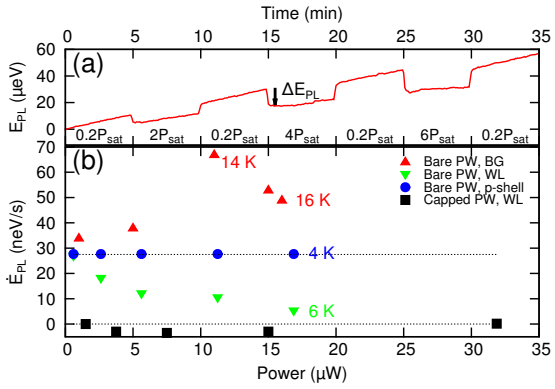

FIG. 3. (Color online)(a) E_{PL} of a QD versus time for various WL excitation power (bare PW, T =4 K) (b) $\dot{E}_{\mathrm{PL}}$ versus power of a bare PW for three excitation energies: $\mathrm{E_{BG}}$ =1.52 eV (▲), $\mathrm{E_{WL}}$ =1.49 eV (●) and $\mathrm{E_{pshell}}$ =1.42 eV (▼), and for a capped PW (■). The PW temperature T_{PW} deduced from the shift ΔE_{PL} is indicated next to some points.

Fig. 3(b)). For p-shell excitation, where no free carrier is injected in the PW and no heating is generated, it remains constant. In the case of WL excitation, where T_{PW} remains below 6 K, one observes a decrease of $\dot{E}_{\mathrm{PL}}$. On the other hand for BG excitation, T_{PW} at high power is close to the maximum drift temperature of Fig. 2. This explains why we observe an increase of $\dot{E}_{\mathrm{PL}}$, which is dominated by temperature effect. However it is worth noting that its maximum value is 7 times lower than the one predicted by the low excitation results of Fig. 2. Hence it is legitimate to think that, as in the case of WL excitation, the effect of temperature is strongly inhibited by an increase of excitation power. A possible explanation for our observations is the presence of photocreated carriers that screen the surface built-in electric field. This phenomenon should lead to a dramatic reduction of the electric field seen by the QD and hence induce a blue shift of its emission energy. This is in direct contradiction with the results of Fig. 3(a). A another explanation that remains to be confirmed could be a direct influence of the carriers or excitation light on the dynamics of adsorption and desorption of O_2 or on its capture of an electron.

The power dependence of $\dot{E}_{\mathrm{PL}}$ for capped PWs is given in Fig. 3(b) and is consistent with a complete suppression of the drift. Although there may still exist a static electric field inside the PW due to surface states at the GaAs/Si_3N_4 interface, its value is frozen and no longer depends on the adsorption of molecules on the device. Frequency stability of the emission of a single QD is crucial to perform high precision resonant spectroscopic studies.

In our device, the QD now behaves as a stable two-level system embedded into an optical waveguide made by the PW. It is an ideal situation for studying non-linear effects at the single photon level and implementing simple quantum information operation between light and a solid state emitter[1].

We acknowledge fruitful discussions with O. Demichel and M. Richard. This work is supported by ANR (P3N project CAFE) and by fundation "Nanosciences aux limites de la Nanoélectronique". Sample fabrication was done in the PTA and CEA LETI MINATEC/DOPT clean rooms.

REFERENCES

[1] A. Auffèves-Garnier, C. Simon, J.-M. Gérard, and J.-P. Poizat, Phys. Rev. A **75**, 053823 (2007).

[2] D. Englund, A. Faraon, I. Fushman, N. Stoltz, P. Petroff, and J. Vuckovic, Nature **450**, 857 (2007).

[3] I. Friedler, C. Sauvan, J. P. Hugonin, P. Lalanne, J. Claudon, and J. M. Gérard, Opt. Express **17**, 2095 (2009).

[4] J. Claudon, J. Bleuse, N. S. Malik, M. Bazin, P. Jaffrennou, N. Gregersen, C. Sauvan, P. Lalanne, and J.-M. Gérard, Nat Photon **4**, 174 (2010).

[5] T. M. Babinec, H. J. M., M. Khan, Y. Zhang, J. R. Maze, P. R. Hemmer, and M. Loncar, Nat Nano **5**, 195 (2010).

[6] J. Bleuse, J. Claudon, M. Creasey, N. S. Malik, J.-M. Gérard, I. Maksymov, J.-P. Hugonin, and P. Lalanne, Phys. Rev. Lett. **106**, 103601 (2011).

[7] O. Demichel, M. Heiss, J. Bleuse, H. Mariette, and A. F. i Morral, Appl. Phys. Lett. **97**, 201907 (2010).

[8] Y. Dan, K. Seo, K. Takei, J. H. Meza, A. Javey, and K. B. Crozier, Nano Letters **11**, 2527 (2011).

[9] S. Tanaka, S. D. More, J. Murakami, M. Itoh, Y. Fujii, and M. Kamada, Phys. Rev. B **64**, 155308 (2001).

[10] P. W. Fry, I. E. Itskevich, D. J. Mowbray, M. S. Skolnick, J. J. Finley, J. A. Barker, E. P. O'Reilly, L. R. Wilson, I. A. Larkin, P. A. Maksym, M. Hopkinson, M. Al-Khafaji, J. P. R. David, A. G. Cullis, G. Hill, and J. C. Clark, Phys. Rev. Lett. **84**, 733 (2000).

[11]B. D. Gerardot, S. Seidl, P. A. Dalgarno, R. J. Warburton, D. Granados, J. M. Garcia, K. Kowalik, O. Krebs, K. Karrai, A. Badolato, and P. M. Petroff, Appl. Phys. Lett. **90**, 041101 (2007).

[12]Y. Chen, F. Stepniak, J. M. Seo, S. E. Harvey, and J. H. Weaver, Phys. Rev. B **43**, 12086 (1991).

[13]H. Nienhaus and W. Mönch, Appl. Surf. Sci. **65**, 632 (1993).

[14]S. G. Anderson, T. Komeda, J. M. Seo, C. Capasso, G. D. Waddill, P. J. Benning, and J. H. Weaver, Phys. Rev. B **42**, 5082 (1990).

[15]R. E. Honig and R. O. Hook, RCA Rev. **21**, 360 (1960).